Usage Report for Fiscal Year 2017

Project Title: First-principles studies on electronic structures of magnetic pyrochlore oxides $A_2Ir_2O_7$ and Ir_2O_4

Name: Shigeki Onoda

Laboratory at RIKEN: Condensed Matter Theory Laboratory

---

1. This project was taken over from the previous ones (Q15307 and Q16307), where strongly correlated pyroochlore iridates $R_2Ir_2O_7$ and A-site-deintercalated iridium spinel oxides Ir_2O_4 were systematically studied using first-principles calculations, in particular, based on the pseudo-atomic orbital and pseudo-potentials implemented into OpenMX package. The previous calculations successfully explained experimental findings on the phase transition from a paramagnetic semimetal for R=Pr through a possible all-in, all-out antiferromagnetic Weyl semimetal to an all-in, all-out antiferromagnetic Mott insulator for R=Nd, Sm, Eu, and Y in $R_2Ir_2O_7$ and uncover unprecedented results on a possible bosonic U(1) quantum spin liquid state in Mott-insulating tetragonal Ir_2O_4 on a particular substrate. The former work was already published, while the manuscript for the latter was submitted but received some requests for additional calculations in the referee process. This year, we have aimed at accomplishing these additional calculations by using another package, ELK, for all-electron first-principle calculations, to reconfirm the previous findings obtained by OPENMX.

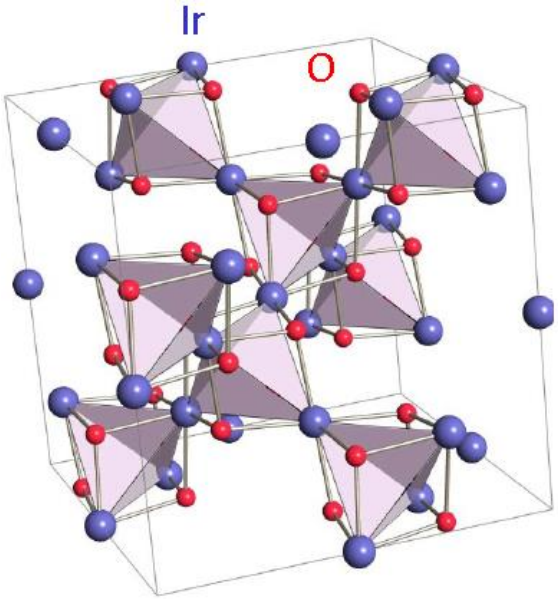

Fig.1: Crystal structure of the ideal cubic A-site-deintercalated spinel iridate Ir_2O_4.

2. We employ first-principles electronic structure calculations based on the local spin density functional with the onsite Coulomb interaction U and relativistic corrections, in particular, using the ELK package. We have used roughly 860k hours of CPU time on the MPC system and 65k hours on the ACSG system by Feb. 15, occupying 3 to 32 nodes for each computational job.

3. We have investigated the ideal cubic case of Ir_2O_4 by the ELK package for the LSDA+U calculations. The results have then been compared with those obtained by the OPENMX package. Figures 2(a) present the previously obtained OPENMX result on the comparison among several (meta)stable states labeled by the magnetic space group. As we increase U, the system undergoes a couple of phase transitions from a metallic paramagnet ($Fd\bar{3}m1'$) through a metallic splayed

ferromagnet ($I4_1/am'd'$) to a Mott-insulating antiferromagnet ($I4_1'/amd'$). Figure 2(b), on the other hand, gives the energy difference between two newly obtained low-energy magnetically ordered states ($I4_1/am'd'$ and $I4_1'/amd'$) obtained with the ELK package in a moderately correlated (intermediate-U) regime. Except that the value of U strongly depends on the basis wavefunctions taken in first-principles calculation packages, the tendency that a moderately large U triggers a phase transition from the metallic splayed ferromagnet to the Mott-insulating antiferromagnet has been confirmed successfully.

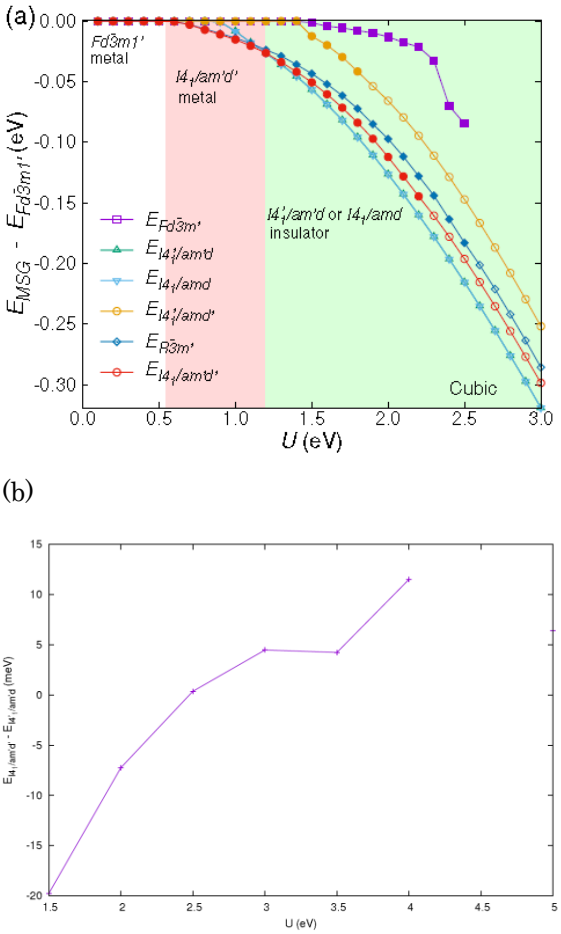

Fig.2: (a) OPENMX results on the energies of (meta)stable magnetically ordered states compared to the paramagnetic state as functions of U, in the ideal cubic case. (b) ELK results on the energy difference between the two magnetically ordered states ($I4_1/am'd'$ and $I4_1'/amd'$)

4. Our complemental all-electron first-principles calculations using the ELK package have revealed that A-site-deintercalated spinel iridates Ir_2O_4 are candidates for quantum spin ice with a spin-ice-rule coupling that is two orders of magnitude larger than in magnetic rare-earth pyrochlores, for instance, $Pr_2Zr_2O_7$, $Tb_2Ti_2O_7$, and $Yb_2Ti_2O_7$.

5. Calculations for understanding the latest experimental reports in $(R_{1-x}Ca_x)_2Ir_2O_7$ and the magnetic neutron-scattering diffraction pattern of $R_2Ir_2O_7$ are still on the way. This study requires a serious implementation of the magnetic structure factor into first-principles calculation packages. We plan to incorporate this issue with a Genera Use project on frustrated magnets that is applied for the next fiscal year.

Fiscal Year 2017 List of Publications Resulting from the Use of the supercomputer

Shigeki Onoda, Fumiyuki Ishii, "First-principles design of the iridate spinel Ir_2O_4 for high-temperature quantum spin ice", arXiv:1612.00553.

[Oral presentation at an international symposium]

Shigeki Onoda, "Quest to U(1) quantum spin liquids, valence bond solids, and novel ordered phases in pyrochlores and spinels: unconventional quasiparticles and interference effects", Junjiro Kanamori Memorial International Symposium (Univ. of Tokyo, Tokyo, Sep. 27-29, 2017). (Invited.)

Shigeki Onoda, "Quest to U(1) quantum spin liquids, valence bond solids, novel ordered phases in pyrochlores and spinels: unconventional quasiparticles and interference effects", Topological States and Phase Transitions in Strongly Correlated Systems (Kavli Institute for Theoretical Sciences, Univ. of Chinese Academy of Sciences, Jul. 3-21, 2017). (Invited.)

Shigeki Onoda, "Quantum spin ice" Part I, II, and III, Frustrated Magnetism: School (Institute of Mathematical Sciences (IMSc), Chennai, India, Apr. 3-9, 2017). (Invited.)